TITLE:	**PALLADIUM/COPPER ALLOY COMPOSITE MEMBRANES FOR HIGH TEMPERATURE HYDROGEN SEPARATION**
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GRANT NUMBER:	DE-FG-03NT41792
PERIOD OF PERFORMANCE:	June 9, 2004 – June 6, 2005
DATE:	March 25, 2005

OBJECTIVES

The specific objectives that we have pursued during the last year are:

- To identify the structural changes that electrolessly deposited palladium alloy films on porous supports undergo when exposed to gas mixtures at a range of temperature and pressure conditions with the purpose of determining the effect of non-metallic impurities such as carbon, and

- To determine the permeation properties and physical and mechanical resistances of these membranes when exposed to gaseous mixtures containing such gases as CO, CO_2, H_2O and H_2S at different compositions,

ACCOMPLISHMENTS TO DATE

Sequential layers of palladium and copper metals were electrolessly deposited on porous Al_2O_3 and ZrO_2 ceramic micro and ultrafilters. The composite membranes thus made were annealed and tested at temperatures ranging from 250 to 500 °C, under very high feed pressures (up to 450 psig) using pure gases and gaseous mixtures containing H_2, CO, CO_2, H_2O and H_2S, with the purpose of determining the effects these variables had on the H_2 permeation rate, selectivity and percent recovery. The inhibition caused by CO/CO_2 gases on a 7 μm thick Pd-Cu composite membrane was less than 17% over a wide range of compositions at 350 °C for CO concentrations up to 8 mole %. The sum of the CO and CO_2 compositions was held constant at 28 mole %. H_2S caused a strong inhibition of the H_2 flux of the same Pd-Cu composite membrane, which is accentuated at levels of 100 ppm or higher. The membrane was exposed to 50 ppm three times without permanent damage. At higher H_2S levels, above 100 ppm the membrane suffered some physical degradation and its performances was severely affected. The use of sweep gases improved the hydrogen flux and recovery of a Pd-Cu composite membrane.

Recently, we have been able to dramatically reduce the thickness of these Pd alloy membranes to approximately one micron. This is significant because at this thickness, it is the cost of the porous support that controls the materials cost of a composite Pd alloy membrane, not the palladium inventory. Very recent results show that the productivity of our membranes is very high, essentially meeting the DOE pure hydrogen flux target value set by the DOE Hydrogen Program. These results were obtained at 365 °C and a differential pressure of 20 psig when a 1.3-micron-thick $Pd_{95}Cu_5$ (composition given in mass %) alloy film was coated on a Pall Corporation Membralox® T1-70 tubular ceramic substrate. This is significant because at this thickness, it is the cost of the porous support that controls the materials cost of a composite Pd alloy

membrane, not the palladium inventory. The H_2 permeability of this membrane was higher than that of our previous work due to reduction of the carbon content of the Pd alloy film. The productivity of this membrane would be even **higher** if the alloy composition was 40 wt. % Cu.

FUTURE WORK

- Continue working with pure SS and modified SS supports in order to duplicate the performance of Pd alloy membranes supported on porous ceramic filters,

- Improve the quality and reproducibility of our deposition techniques to minimize impurities and produce Pd alloy membranes containing the highest performance 40 weight % copper concentration.

LIST OF PAPERS PUBLISHED, CONFERENCE PRESENTATIONS, STUDENTS SUPPORTED

JOURNAL ARTICLES AND CONFERENCE PROCEEDINGS

Kulprathipanja, A., Alptekin, G. O., Falconer, J. L. and J. D. Way, "The Effects of Water Gas Shift Gases on Pd-Cu Alloy Membrane Surface Morphology and Separation Properties," *Ind. Eng. Chem. Res.*, **43**(15), 4188-4198(2004).

Roa, F., Way, J. D., DeVoss, S., and G. Alptekin, "Effect of CO_2, CO, H_2S on a Pd-Cu Ceramic Composite Membrane Made by Electroless Plating", Proceedings of the International Conference on Inorganic Membranes (ICIM 8), Cincinnati, OH, July 18-22, 2004.

Roa, F., Way, J. D., DeVoss, S., and G. Alptekin,, "Thin Palladium Copper Membranes for Hydrogen Separation," Proceedings of the World Renewable Energy Conference (WREC), Denver, CO, August 30 - September 3, 2004.

Roa, F. and J. D. Way, "The Effect of Air Exposure on Palladium-Copper Composite Membranes," *Applied Surface Science*, **240**(1-4), 85-104(2005).

Kulprathipanja, A., Alptekin, G. O., Falconer, J. L., and J. D, Way, "Pd and Pd-Cu Membranes: Inhibition of H_2 Permeation by H_2S, "*Journal of Membrane Science*, in press, 2005.

Thoen, P. M., Roa, F., and J. D. Way, "High Flux Palladium-Copper Composite Membranes for Hydrogen Separations," submitted to *Desalination*, 3/2005.

CONFERENCE PRESENTATIONS

Roa, F., Way, J. D., DeVoss, S., and G. Alptekin, "Effect of CO_2, CO, H_2S on a Pd-Cu Ceramic Composite Membrane Made by Electroless Plating", Paper presented at the International Conference on Inorganic Membranes (ICIM 8), Cincinnati, OH, July 18-22, 2004.

Roa, F., Way, J. D., DeVoss, S., and G. Alptekin,, "Thin Palladium Copper Membranes for Hydrogen Separation," Paper presented at the World Renewable Energy Conference (WREC), Denver, CO, August 30 - September 3, 2004.

STUDENTS SUPPORTED

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